Looking at the superconducting gap of iron pnictides

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Abstract

THz and infrared spectroscopy is widely utilized to investigate the electrodynamic properties of the novel iron-based superconductors in the normal and superconducting states. Besides electronic excitations and correlations, electron-phonon coupling and the influence of magnetism, the experiments yield important information on low-lying excitations and help to clarify the number and symmetry of superconducting gaps. While the experimental data of different groups converge, the interpretation is still under debate. Here we review the status of optical investigations on the superconducting state for the 122 and 11 family of iron pnictides.

Keywords: Superconducting energy gap, Iron pnictides, Optical spectroscopy, Order parameter
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1. Introduction

Macroscopically superconductivity is characterized by vanishing resistivity and perfect diamagnetism; microscopically, a gap in the density of states opens at the Fermi surface [1]. Both phenomena are subject of the BCS theory which assigns an order parameter to the superconducting state. This immediately gives some insight into the symmetry and eventually the mechanism of superconductivity [2]. It works almost perfectly in conventional single band metallic superconductors, it has some success in the description of more exotic systems [3], such as heavy fermions [4] or organic superconductors [5], but requires care when applied to high-temperature superconductivity in transition metal oxides.

When the new class of iron pnictides was discovered two years ago [6], immediately the questions of mechanism and symmetry arose. Despite the enormous efforts undertaken of the entire community [7] the present answers are not completely satisfactory.

Since the development of the BCS theory, optical investigations contributed a great deal to solve the puzzle of superconductivity. And for each new family of exotic compounds discovered over the decades, high-frequency studies and infrared spectroscopy were one of the first and decisive investigations frequently pointing the direction for further studies of complementary methods [8, 9, 10, 11].

Following these lines, optical methods were quickly applied to iron pnictides and revealed the spin-density-wave gap in parent compounds, as well as the electronic properties of the superconducting systems [12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24].

By now there is only a limited number of optical investigations on the superconducting properties of iron-pnictides. Early studies on non-crystalline materials gave some rough ideas [12, 25, 26, 27, 28], but could not contribute to the main questions posed above. Only very recently, several groups published their findings on single crystals of the 122 family [16, 29, 30, 31, 32] and 11 compound [33]. Also epitaxial grown thin films have been explored [34, 35]. As we will review in this contribution, the optical data look very similar, but interestingly, the interpretation is very dispersed.

2. Results and Discussion

2.1. Hole-doped 122 iron pnictides

In very early experiments on the hole-doped compound $Ba_{0.6}K_{0.4}Fe_2As_2$, Li *et al.*[16] found clear indications of the opening of a complete gap. The reflectivity shows a sharp upturn below T_c which approaches unity around 150 cm^{-1}; the optical conductivity drops to zero in a linear fashion as demonstrated in Fig. 1. The behavior can be fitted by assuming a single *s*-wave gap with $2\Delta_0/hc = 110$ cm^{-1}.[1] A fit by two gaps ($2\Delta_0^{(2)}/hc = 110$ cm^{-1} and $2\Delta_0^{(1)}/hc = 190$ cm^{-1}) gives a slightly better description [18]. From the spectral weight removed below the gap, the superconducting penetration depth of $Ba_{0.6}K_{0.4}Fe_2As_2$ can be calculated: $\lambda = 2000$ Å. The spectral weight analysis shows that the Ferrell-Glover-Tinkham sum rule [1, 36] is satisfied below $6\Delta_0$.

2.2. Electron-doped 122 iron pnictides

2.2.1. Films

Comprehensive optical investigation of $Ba(Fe_{0.9}Co_{0.1})_2As_2$ thin films in a wide frequency and temperature range elucidate

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[1] Here $h = 2\pi\hbar = 4.1\times10^{-15}$ eV s is the Planck constant, and $c = 3\times10^8$ m/s is the speed of light. Note that optical experiments probe excitations across the full gap of 2Δ.

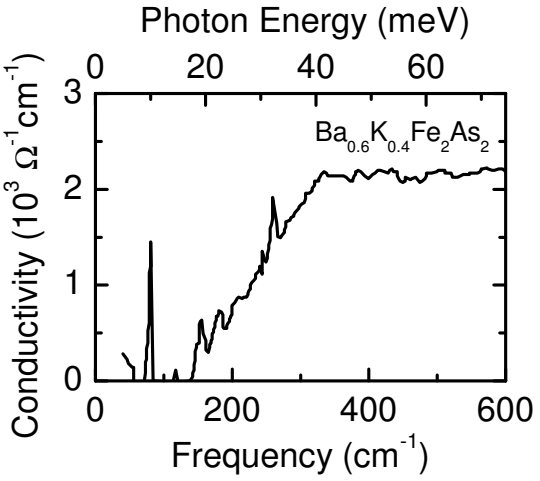

Figure 1: Frequency dependent conductivity of $Ba_{0.6}K_{0.4}Fe_2As_2$ obtained from reflectivity measurements at $T = 10$ K. The data are taken from Li *et al.* [16].

the electrodynamic properties in the superconducting state. A 900 Å thick film with $T_c = 20$ K was laser-deposited on a (La,Sr)(Al,Ta)O_3 substrate. Using a Mach-Zehnder interferometer Gorshunov *et al.* [34] were able to directly observe the opening of the superconducting gap of $2\Delta_0 = 3.7$ meV corresponding to 30 cm^{-1}, i.e. $2\Delta_0/k_BT_c = 2.1 \pm 10\%$ by THz transmission and phase shift measurements. In this region the temperature and frequency dependence of the conductivity is well described by the BCS theory assuming a complete isotropic gap, as plotted in Fig. 2(a). However, there remains a strong quasi-particle absorption below 10 cm^{-1}, which is not predicted by the simple model. The spectral weight of the condensate 1.94×10^7 cm^{-2} corresponds to a penetration depth $\lambda = 3600$ Å.

Using synchrotron radiation, Perucchi *et al.* measured the THz reflectivity of a high quality epitaxial thin film of Ba$(Fe_{0.92}Co_{0.08})_2As_2$ (thickness $d = 3500$ Å with $T_c = 22.5$ K) on a $DyScO_3$ substrate with an epitaxial $SrTiO_3$ intermediate layer [35]. They did not evaluate the intrinsic material properties, but only the composite reflectance of film and substrate, and presented the ratio of superconducting and normal states: $R_s(T)/R_n$. For that reason no conductivity could be calculated and plotted here. Nevertheless, clear evidence for a superconducting gap $2\Delta_0^{(1)}/hc \approx (30 \pm 1)$ cm^{-1} was observed. Following the approach of other groups and methods, a fit by a two-band, two-gap model yields better agreement. The larger gap is estimated to be at approximately $[2\Delta_0^{(2)}/hc = (110\pm15)\,\text{cm}^{-1}]$. The results are seen as evidence for a nodeless isotropic double-gap scenario, with the presence of two optical gaps corresponding to $2\Delta_0/k_BT_c$ values close to 2 and 7. The two contributions to the optical conductivity have a plasma frequency of 10^4 cm^{-1} and scattering rates of 300 and 1200 cm^{-1}, respectively.

2.2.2. Single Crystals

Reflectivity measurements on single crystals suffer from small crystal size and limited surface quality. Nevertheless it is quite remarkable how well the data of different groups coincide, as demonstrated in Fig. 2(b) which shows the optical conductivity as obtained by Kramers-Kronig analysis as a function of frequency for the lowest temperature available. In all cases significant deviations from the normal state behavior occur below 200 cm^{-1}, which corresponds to approximately $6 - 8\Delta_0$. In the superconducting phase the conductivity passes through a maximum around 80 cm^{-1}, before it drops. There seems to be a considerable in-gap absorption present as a more gradual reduction with decreasing frequency than expected for a simple *s*-wave superconductor.

The first broadband investigations on single crystals of electron-doped iron pnictides were performed by Wu *et al.* [29, 30]. They measured Ba$(Fe_{0.92}Co_{0.08})_2As_2$ in a wide temperature range and frequencies from 20 cm^{-1} to 30 000 cm^{-1}. Upon passing the superconducting transition at $T_c = 25$ K, the reflectivity rises toward unity, leading to a clear gap-like feature in the conductivity as depicted in Fig. 2(b) as a solid line. The conductivity below 100 cm^{-1} drops due to the complete opening a gap in the density of states at $2\Delta_0^{(1)}/hc = 50$ cm^{-1}, corresponding to $2\Delta_0/k_BT_c \approx 2.5 - 3$. It can be well described by the BCS theory with no nodes assumed in the order parameter. There remains a considerable contribution below which infers that not all carriers are gapped. Alternatively, a superior fit is suggested with a second gap at 17 cm^{-1}, following suggestions by APRES that two gaps are present [38, 39, 40]. The missing spectral weight amounts to a penetration depth of $\lambda = (3500 \pm 350)$ Å for Ba$(Fe_{0.92}Co_{0.08})_2As_2$.

The behavior of Ba$(Fe_{0.95}Ni_{0.05})_2As_2$ is very similar to the optimally doped Co samples [29, 30]. The reflectivity turns up around 60 cm^{-1} and approaches unity in way that the optical conductivity becomes gapped with $2\Delta_0^{(1)}/hc = 35$ cm^{-1}. In Fig. 2(c) the frequency dependent conductivity is plotted for $T = 10$ K, i.e. well into the superconducting state ($T_c = 20$ K). The larger carrier density leads to a slightly smaller penetration depth of $\lambda = (3000 \pm 300)$ Å.

Nakajima *et al.* presented a comprehensive study of several Ba$(Fe_{1-x}Co_x)_2As_2$ compounds with $x = 0$, 0.04, 0.06, and 0.08, where the latter two become superconducting at $T_c = 25$ and 20 K [21]. The normal state conductivity is very well described by two Drude components with very different scattering rates, as suggested by Wu *et al.* [29]. The narrow Drude term becomes stronger in spectral weight with increasing the Co concentration *x*; the broad incoherent background is assigned to the segments of the Fermi surface which fulfill the nesting conditions. From the rise in reflectivity and the corresponding suppression of $\sigma_1(\omega)$ at the lowest temperature ($T = 5$ K), they conclude a full gap superconductor of *s*-wave symmetry with $2\Delta/hc \approx 80(50)$ cm^{-1}, which yields $2\Delta/k_BT_c = 4.6$. No conclusion can be drawn about a second gap above or below this major feature. As pointed out, the gap seems to develop in every piece of the Fermi surface in the superconducting state since the narrow and the broad Drude term are equally affected by the superconducting gap. From the missing area, the penetration depth $\lambda = (2770 \pm 250)$ and (3150 ± 300) Å is estimated, for the two compounds.

Temperature dependent reflectivity measurements on Ba$(Fe_{1-x}Co_x)_2As_2$ were also performed by van Heumen *et al.* [32]. In Fig. 2(b) we present the data obtained on Ba$(Fe_{0.93}$-$Co_{0.07})_2As_2$ at $T = 10$ K as dotted line. In contrast to other

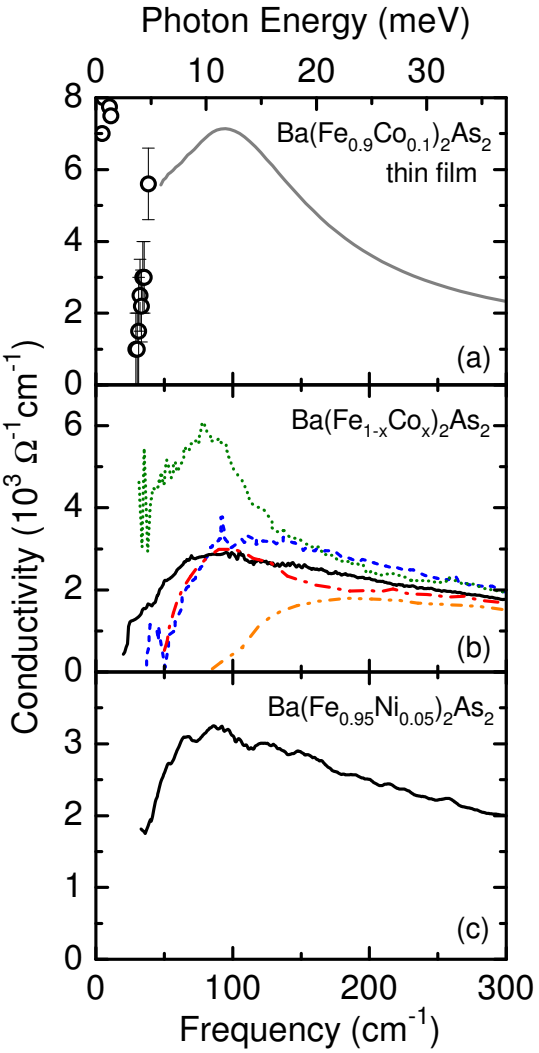

Figure 2: Optical conductivity of electron doped $Ba(Fe_{1-x}M_x)_2As_2$ close to optimal doping; M=Co, Ni. (a) From THz transmission and phase measurements through a thin film of $Ba(Fe_{0.9}Co_{0.1})_2As_2$ at $T = 5$ K (open dots) the complete opening of the gap in conductivity can be seen around 30 cm^{-1}; the grey line is obtained by reflectivity measurements. (b) Reflectivity measurements performed by different groups yield a very similar behavior at low temperatures. The solid line was obtained by Wu *et al.* [29, 30] on $Ba(Fe_{0.92}Co_{0.08})_2As_2$ at $T = 9$ K. Kim *et al.* [31] investigated $Ba(Fe_{0.935}Co_{0.065})_2As_2$ at $T = 5$ K (blue dashed curve). The green dotted curve corresponds to a measurement of van Heumen *et al.* [32] on $Ba(Fe_{0.93}Co_{0.07})_2As_2$ at $T = 10$ K. Nakajima *et al.* [21] investigated $Ba(Fe_{0.94}Co_{0.06})_2As_2$ (dot-dashed red line) and $Ba(Fe_{0.92}Co_{0.08})_2As_2$ (dot-dot-dashed orange line) single crystals with $T_c = 25$ and 20 K, respectively. (c) The behavior of the $Ba(Fe_{0.95}Ni_{0.05})_2As_2$ at $T = 10$ K is very similar to the Co doped sister compound [29].

groups, they find a strong peak around 80 cm^{-1} in the conductivity spectrum which is assigned to interband transitions; this makes the separation of the free charge carrier conductivity difficult. In the course of their analysis of the missing area the superfluid density was estimated to $(2.2 \pm 0.5) \times 10^7$ cm^{-2}. Plotting $-\omega^2\epsilon_1(\omega)$ shows a minimum around 50 cm^{-1}, which indicates a gap of $2\Delta_0^{(1)} \approx 6.2$ meV with s-wave symmetry [41]. Although no other gap structure is apparent in the data, van Heumen *et al.* decomposed the conductivity in a approach similar to [29] and found them to be consistent with a second gap around $2\Delta_0^{(2)} = 14$ meV as suggest by ARPES and STS experiments [42, 43].

The optical properties of Co-doped $BaFe_2As_2$ were also measured by Kim *et al.* [31] at temperatures $T \geq 5$ K. The conductivity of a single crystal of $Ba(Fe_{0.935}Co_{0.065})_2As_2$ ($T_c = 24.5$ K) as derived by Kramers-Kronig analysis of reflectivity data down to 35 cm^{-1} is plotted in Fig. 2(b) as dashed line. In the normal state two Drude contributions [with $1/(2\pi c\tau) = 90$ and 300 cm^{-1}, respectively] and some mid-infrared Lorentzians are sufficient to describe the frequency-dependent conductivity; in agreement to the approach the other groups have chosen. Also in the superconducting state their findings are in accord with previous experiments by Wu *et al.* [29] (solid line) as far as the overall behavior is concerned. However, Kim *et al.* point out that a multi-gap scenario with at least three energy gaps ($2\Delta/k_BT_c = 3.1$, 4.7 and 9.2) is necessary to reproduce the experimental data. While the drop of $\sigma(\omega)$ to zero determines the gap $2\Delta_0^{(1)}/hc = 53$ cm^{-1} unambiguously, it is hard to identify gaps at 80 and 157 cm^{-1} in their raw data. There remains a strong in-gap absorption which becomes more pronounced at $T = 20$ K and by far exceeds the predictions of a complete s-wave scenario. While the gap $\Delta^{(1)}$ opens in the narrow Drude, the 80 cm^{-1} gap is assigned to the broad Drude contribution. It is interesting to note the common trend, that the larger the gap is, the smaller the spectral weight that contributes to the corresponding conductivity. The penetration depth $\lambda = 2700$ Å is estimated in their report.

In summary, as already pointed out in Ref. [35], most groups measuring reflectivity of electron-doped single crystals agree on a 50 cm^{-1} gap,[2] while there is some discrepancy on the existence of further gaps. On the other hand, both investigations carried out on thin films indicate a gap of $2\Delta/hc \approx 30$ cm^{-1}. In trying to explain this disaccord, disorder or strain effect have to be considered. Furthermore, it is worth to note that, in all cases the further gap(s) favor to open in individual conducting channels where the scattering rate is rather big. Thus, a universal decomposition of free carrier contribution corresponding to the Fermi-surface construction is aspired to get deeper insight of the gap analysis.

2.3. *$FeTe_{0.55}Se_{0.45}$ compound*

The so-called 11 compounds are the simplest in the iron pnictide family, and the As-free FeSe reaches $T_c = 8$ K which

[2] The rather large gap value reported for the hole-doped compound [16] waits for an independent confirmation.

can be increased to 27 K upon application of pressure. By introducing Te, the critical temperature reaches a maximum of $T_c = 14$ K for in $FeTe_{0.55}Se_{0.45}$. Homes *et al.* have explored the electrodynamic properties by performing reflectivity measurements down to 6 K [33]. Very similar to the 122 compounds, an almost constant conductivity background is observed in the range between 500 and 1000 cm^{-1} on top of which a Drude-like component develops as the temperature is reduced. The authors fit their data by the Drude-Lorentz approach [36] with a strong mid-infrared excitation which contains a considerable tail all the way down to the lowest frequencies. In the superconducting state at $T = 6$ K the optical conductivity is reduced below 120 cm^{-1} with a prominent shoulder at 60 cm^{-1} as plotted in Fig. 3. Below the maximum in $\sigma(\omega)$ the conductivity drops in a linear way towards zero. It is interesting to note that only a quarter of the free charge carriers collapse into the condensate leading to $\lambda = 5300$ Å. Although a Drude fit of the normal state yields $1/(2\pi c\tau) \approx 30$ cm^{-1}, the material is considered in the dirty limit as the frequency dependent scattering rate $1/\tau(\omega)$ is enlarged in the region of the gaps.

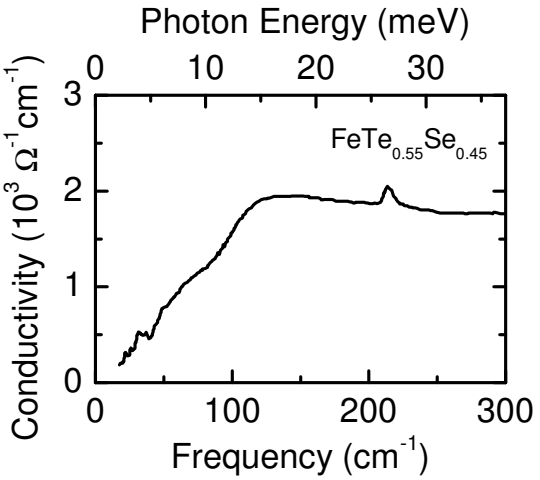

Figure 3: Optical conductivity of a $FeTe_{0.55}Se_{0.45}$ single crystal ($T_c = 14$ K) obtained from reflectivity measurements at $T = 6$ K by Homes *et al.* [33].

The data can be fitted by the Mattis-Bardeen expression [37] using a single gap of $2\Delta_0 = 10.2$ meV (corresponding to 82 cm^{-1}), but the remaining low-lying excitation call of a two-gap fit with $2\Delta_0^{(1)} = 5$ meV and $2\Delta_0^{(2)} = 10.2$ meV, very similar to the approach Wu *et al.* used in the electron doped 122 compounds [29]. Some in-gap excitations remain, far more than expected from a simple *s*-wave gap at temperatures below $T_c/2$. Nevertheless Homes *et al.* conclude that the energy gaps in this material are isotropic and see no indications of nodes in the order parameter [33]. The different gaps belong to different bands crossing the Fermi surface, although a clear assignment cannot be made from optical experiments. Since the gaps open simultaneously with decreasing temperature below $T_c = 14$ K, the coupling constants are different $2\Delta_0/k_BT_c = 4$ and 8.4.

3. Comparison with MgB_2

Is is worth to have a closer look at optical measurements on MgB_2, which is a well-known two-band superconductor [44]. In general a gap value corresponding to the π bands of approximately $2\Delta_\pi/hc = 25 - 40$ cm^{-1} is extracted [45, 46, 47, 48, 49], corresponding to $2\Delta_\pi/k_BT_c \approx 1 - 2$. However, these infrared reflection and transmission experiments could not see any indication of the larger $2\Delta_\sigma$, expected around 110 cm^{-1} based on tunneling [51], ARPES [52], and Raman [53] investigations. Only recently, measurements on thin films with low and high impurity levels give some indications of two gaps in the optical response [50]. Commonly the infrared data are analyzed by assuming that the charge carriers in the π and σ bands respond independently to the external radiation; each component is characterized by its own plasma frequency ω_p, scattering rate $1/\tau$ and superconducting gap 2Δ. Although a suitable choice of parameters allows one to fit the data, the description remain unsatisfactory because a rise in reflectivity should always occur at the value of the larger gap.

4. Conclusion and Outlook

The comparison of the various optical results on single crystals and thin films of iron pnictides give a very similar overall picture. The optical conductivity in the normal state consists of some mid-infrared bands and a quasi-free carrier contribution at $\omega = 0$, which cannot be described by one simple Drude term. Frequently the optical conductivity is decomposed into two independent contributions (except for the FeSe compound where only one Drude was used), albeit no obvious assignment is possible to the bands crossing the Fermi surface. Entering the superconducting state the conductivity drops as the electronic density of states becomes gapped.

No agreement exists as far as the number of gaps is concerned. Mainly based on suggestions from other experimental methods and theory, gaps of different size are tested. The common approach is that simultaneously below T_c, each of the conductivity contributions become completely gapped with different coupling strength $2\Delta/k_BT_c$. Care has to be taken not to overinterprete the conductivity spectra above 100 or 200 cm^{-1} since correlation and bandstructure effect as well as phonons might cause deviations form a simple Drude behavior.

Looking in more detail at the optical conductivity obtained by different groups for $T < T_c$, however, significant deviations become obvious, in particular at the low-frequency end of the spectrum. By now, only simple *s*-wave scenarios have been considered with no *k*-dependent order parameter. In a detailed analysis of the optical conductivity of iron pnictides, Carbotte and Schachinger [54] pointed out that in an extended *s*-wave scenario gap nodes may exist in certain directions on the Fermi surface. These nodes can be lifted by increasing disorder leading to a finite gap in all momentum directions. Further low-frequency experiments have to be performed, well below 50 cm^{-1}, in order to clarify this issue.

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